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Thermodynamic properties of concentrated spin glasses: A cluster mean-field theory

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Using a previously discussed cluster mean-field theory we compute the static properties for the concentrated spin glasses. These are viewed as alloys in which the average cluster size is large. We find for these large clusters that, with an Ising ferromagnetic intracluster Hamiltonian, the static susceptibility χ has a cusp at the critical temperature T_c while the specific heat C_m has a rounded maximum at a higher temperature T_o. These results are in accordance with those previously obtained for small antiferromagnetic clusters and with experiment. It therefore appears that the cluster mean-field model gives results consistent with the measured thermodynamic properties for both the dilute and concentrated spin-glass regimes.

Recently we proposed a simple cluster mean-field theory (CMFT)[1] of the spin-glasses, in which correlated clusters rather than individual spins are the basic entity. Starting with a Heisenberg Hamiltonian with random Gaussian-distributed exchange interactions between clusters we computed the static properties of a spin glass. It was found that for a range of average cluster sizes N $(3 \leq N \leq 6)$ and for antiferromagnetic intracluster interactions the CMFT yielded a sharp cusp in the static susceptibility χ at the critical temperature T_C and a rounded maximum in the specific heat C_m at a higher temperature T_0 (which was characteristic of the intracluster exchange energy J_0). Both features are in semiquantitative agreement with experiment. For ferromagnetic intracluster interactions we found that agreement with experiment was not as satisfactory as for small clusters $(N \leq 6)$. The purpose of the present paper is to show that for large N (as is appropriate to the more concentrated spin glasses) agreement between theory and experiment with ferromagnetic intracluster exchange interaction is again obtained. Because we were unable to carry out the numerical calculations with a Heisenberg Hamiltonian for bigger cluster sizes, we compute here the static properties using an Ising Hamiltonian, in which case, we can treat up to 20 spins in the cluster.

We wish to suggest the following physical picture of the spin glasses over the entire range of concentration: For the very dilute alloys N is small and the intracluster interactions are predominantly antiferromagnetic (arising from the RKKY (Rudderman-Kittel-Kasuya-Yosida) interaction). As the alloy concentration increases N increases and for both protypical spin glasses *Au*Fe and *Cu*Mn, the intracluster interaction is predominantly ferromagnetic. [At very high (c >50%) Mn concentration in *Cu*Mn the sign again changes[2] but we will not discuss this limit.] From this point of view, it then becomes possible to understand how the cusp in χ and the broad maximum in C_m coexist for all concentrations up to nearly the percolation limit.

The Ising Hamiltonian

$$H=-\sum_{i,j=1}^{N} J_{ij} S_i S_j$$

can be written in an equivalent form

$$H=-\sum_{i,j=1}^{N_{\text{sp}}} \sum_{\nu,\lambda=1}^{N_{\text{cl}}} J_{i\nu j\lambda} S_{i\nu} S_{j\lambda} ,$$

if we assume that our system has N_{cl} clusters with N_{sp} spins in each cluster and $N=N_{\text{cl}} N_{\text{sp}}$. Splitting this sum into the $\nu=\lambda$ and $\nu \neq \lambda$ terms we have

$$H=-\sum_{\nu<\lambda} J_{\nu\lambda} S_\nu S_\lambda - \sum_{\nu}\sum_{i<j} J_{ij}^0 S_{i\nu} S_{j\nu} , \tag{1}$$

where J_{ij}^0 is the intracluster exchange interactions and $J_{\nu\lambda}$ is the near-neighbor randomly distributed intercluster exchange interactions which now are taken to be centered around a positive value J_1. Greek indices refer to a particular cluster and Roman indices to a given spin within that cluster. Here $S_\nu = \sum_i S_{i\nu}$ and $S_{i\nu} = \pm\frac{1}{2}$. The only assumption we made in writing Eq. (1) is that the clusters are far apart compared to their average size, so $J_{i\nu j\lambda}$ can be taken to be $J_{\nu\lambda}$, i.e., independent of the location of the spins within the clusters. As in the theory of Edwards-Anderson (EA)[3] the intercluster exchange interactions which are given by a near-neighbor (nn) Gaussian distribution

$$P(J_{\nu\lambda}) = (1/\sqrt{2\pi}J) e^{-(J_{\nu\lambda}-J_1)^2/2J^2} \tag{2}$$

are treated within a random mean-field theory. The intracluster interactions are treated exactly.

Using the replica method and following Refs. 1 and 4 we can obtain within the cluster mean-field theory the free energy and from it the self-consistent equations for the three variational parameters M, q, and m. $M=[\langle S_\nu^2\rangle]_c$, where $[\ \ ]_c$ denotes a configurational average, is the total spin of each cluster, $q=[\langle S_\nu\rangle^2]_c$ is analogous to the usual EA spin-glass order parameter and $m=[\langle S_\nu\rangle]_c$ is the long-range ferromagnetic order parameter which is zero in the spin-glass and paramagnetic phases. We find that the free energy per cluster is given by[5]

$$F(q,M)=-kT\left(\frac{\beta^2\bar{J}^2}{4}(q^2-M^2)+\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty}dx\,e^{-x^2/2}\ln \mathrm{Tr}\,e^{-\beta H^{\mathrm{eff}}}\right), \tag{3}$$

where

$$-H^{\mathrm{eff}}=\sum_{i<j}J_{ij}^0 S_{i\nu}S_{j\nu}+\bar{J}\sqrt{q}\,S_\nu x+\tfrac{1}{2}\beta\bar{J}^2(M-q)S_\nu^2. \tag{4}$$

Here $\bar{J}=z^{1/2}J$, with z the number of cluster nn of a given cluster and $\beta=(kT)^{-1}$. The self-consistent equations for the variational parameters are

$$M=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty}dx\,e^{-x^2/2}\frac{\mathrm{Tr}\,S_\nu^2 e^{-\beta H^{\mathrm{eff}}}}{Z}, \tag{5}$$

$$M-q=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty}dx\,e^{-x^2/2}x\frac{1}{\beta\bar{J}\sqrt{q}}\frac{\mathrm{Tr}\,S_\nu e^{-\beta H^{\mathrm{eff}}}}{Z}, \tag{6}$$

where $Z=\mathrm{Tr}\,e^{-\beta H^{\mathrm{eff}}}$. Note that from Eq. (5) we obtain a temperature-dependent cluster moment M; at temperatures high compared to the intracluster exchange interactions $M\to\frac{1}{3}NS(S+1)$ where $S=\frac{1}{2}$ is the spin of a single impurity atom. At $T\to 0$ the cluster moment M has its ground state value ($\frac{1}{4}$ or 0 for perfectly compensated antiferromagnets and N^2S^2 for ferromagnets). Solving Eqs. (5) and (6) analytically near T_C (for the spin-glass-paramagnetic phase), we find that the critical temperature T_C is given by a *self-consistent equation*

$$kT_C/J_0=RM_C, \tag{7}$$

where $R=\bar{J}/J_0$ and M_C is the value of the moment at T_C. The phase diagram is very similar to that found by Sheerington and Kirkpatrick.[6]

The static susceptibility $\chi(T)$[6] is given by

$$\chi(T)=\chi_0(T)/[1-\bar{J}_1\chi_0(T)], \tag{8}$$

where $\chi_0(T)=\beta(M-q)g^2\mu_B^2$ is the result for $J_1=0$ and $\bar{J}_1=zJ_1$. For $\bar{J}_1<\bar{J}$ the effect of $\bar{J}_1\neq 0$ is to enhance the susceptibility in the spin-glass phase. Because of the temperature dependence of M, $\chi(T)$ will differ slightly from its value as obtained in the EA mean-field theory.

The specific heat per cluster C_m is given by

$$C_m=C_m^{\mathrm{inter}}+C_m^{\mathrm{intra}} \tag{9}$$

where the intercluster contribution to the specific heat is

$$C_m^{\mathrm{inter}}=\frac{d}{dT}\left(\frac{\bar{J}^2}{2kT}(q^2-M^2)\right) \tag{10}$$

and the intracluster contribution is given by

$$C_m^{\mathrm{intra}}=\frac{d}{dT}\int_{-\infty}^{+\infty}dx\,e^{-x^2/2}\frac{\mathrm{Tr}(-\sum_{i<j}J_{ij}^0 S_{i\nu}S_{j\nu})e^{-\beta H^{\mathrm{eff}}}}{\sqrt{2\pi}Z}. \tag{11}$$

The intercluster term gives a cusplike contribution at T_C to C_m, similar to that found in the EA mean-field theory. While the intracluster contribution has a rounded maximum at a higher temperature T_0. The specific heat C_m is independent of the J_1, since we are in the spin-glass phase where the long-range ferromagnetic parameter m vanishes.

Because we use an Ising Hamiltonian, it is possible to solve Eqs. (5) and (6) numerically for more intracluster spins (up to 20) than could be done for a Heisenberg model. In this analysis we used 15 spins in a closed configuration (5×3). While in a real spin glass the clusters are not compact but extended, this does not alter the essential conclusions. For the intracluster interactions we consider two cases: First an nn ferromagnetic interaction with a characteristic exchange J_0; in the second case an additional next-nearest-neighbor (nnn) interaction of strength $-0.5J_0$ is also included. The latter is a crude representation of the indirect RKKY interaction between the nnn spins. The ratio of the exchange constant $R=\bar{J}/J_0=0.004$ was chosen so to reduce approximately the experimentally observed ratio for T_C/T_0.

In Fig. 1 are shown the normalized temperature

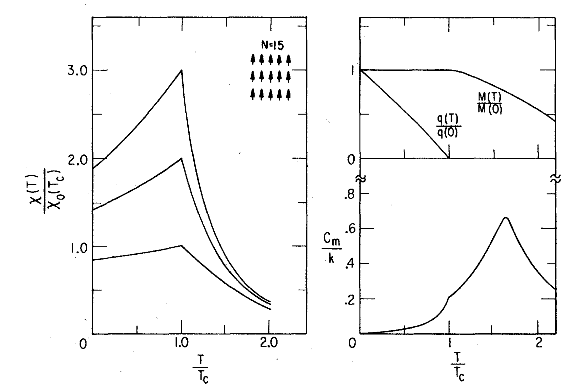

FIG. 1. Temperature dependences of the cluster moment $M(T)$, order parameter $q(T)$, specific heat per spin C_m, and the susceptibility $\chi(T)$. The normalized static susceptibility $\chi(T)/\chi_0(T_c)$ is plotted for $\bar{J}_1/J_0=0.003$, 0.002, 0.0, from top to bottom. The ground-state spin configuration is shown.

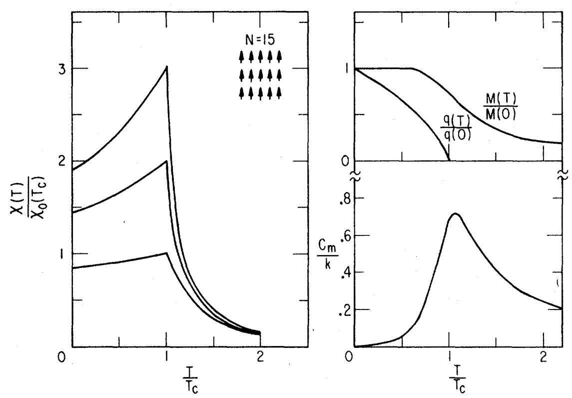

FIG. 2. Temperature dependences of the $M(T), q(T)$ the specific heat per spin C_m, and the susceptibility $\chi(T)/\chi_0(T_c)$ for the same parameter as in Fig. 1 except that a nnn antiferromagnetic between spins within the clusters has been included. The ground-state spin configuration is also shown.

dependence for $M, q, \chi(T)$, and C_m for the first case of nn intracluster ferromagnetic coupling. The order parameter q decreases nearly linearly as a function of increasing temperature as in the EA model. The specific heat has a rounded maximum at a temperature $T_0 > T_C$, while at T_C there remains a small feature which we expect to be washed out by fluctuation effects. Note that the maximum in the specific heat is not so broad as in our previous analysis[1] since we have an Ising Hamiltonian.[7] The susceptibility has a cusp at T_C which is sharper for $\bar{J}_1$ nonzero, similar to that found in EA mean-field theory. For concentrated spin glasses we expect $\bar{J}_1 \neq 0$. This leads to a sharpening in $\chi(T)$ which compares well with experimental results[8] for *Au*Fe.

In Fig. 2 are shown similar results after including both an nn ferromagnetic and nnn antiferromagnetic intracluster coupling. Because of the nnn coupling M decreases more rapidly with temperature. The susceptibility $\chi(T)$ still has a cusp at T_C, but decreases more rapidly above T_C because of the M dependence on temperature. The specific heat has, again, a rounded maximum at a higher temperature than T_C, while there is almost no structure at T_C. It should be noted that Riess[9] has found results similrr to those plotted here using a Green's function decoupling technique which includes fluctuation effects. These fluctuations play a role similar to that of the intracluster interactions.

From the above we derive the following conclusions: (i) The features pertaining to thermodynamic properties we observed for the small antiferromagnetic clusters apply also to the mictomagnetic regime in which the clusters are considerably larger. (ii) With the CMFT we can explain how the cusp in χ and the broad maximum in C_m coexist for all the concentrations up to nearly the percolation limit. From our computer calculations for small antiferromagnetic clusters ($N \leq 6$) we obtain a sharp cusp in $\chi(T)$ at T_C as well as a broad maximum in C_m at a higher temperature T_0. As the number of spins N in the cluster increases ($N \geq 12$) the ferromagnetic clusters also yield a sharp cusp in $\chi(T)$ and broad maximum in C_m. For large antiferromagnetic clusters ($N \geq 12$) we find that the cusp in χ at T_C is washed out. This arises from the unusually large temperature dependence[8] of M which increases from its ground-state value to its infinite temperature result $\frac{1}{3}NS(S+1)$. However, it is important to note that for all large ferromagnetic clusters, this difficulty does not occur and $\chi(T)$ always has a cusp at T_C. This is a consequence of the fact that below T_C, M is almost temperature independent (it is almost constant) because of the small value of $R = \bar{J}/J_0$, which is needed to fit the specific heat, in particular the ratio T_C/T_0. These conclusions are not altered, even when a nnn antiferromagnetic interaction is included.

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